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Origins of Laser Damage in Crystals of KDP

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Final Report

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Introduction

The ability of optical materials to withstand high power ultraviolet (UV) laser irradiation without sustaining irrevocable damage is critically important in two areas central to LLNL: laser fusion and UV lithography. In particular, the output fluence of the National Ignition Facility (NIF) is limited by the 351 nm laser damage thresholds of the KH_2PO_4 (KDP) frequency conversion crystals.[1] The ability to increase the laser output would maximize the odds of achieving ignition, allow target physicists to assess target performance at higher drives, and provide higher temperature-density conditions for studies of the physics of stellar interiors. Moreover, in order to meet the current design criteria for fusion laser systems, KDP crystals must be conditioned by illumination with low fluence laser irradiation to increase the damage threshold by about a factor of two.[2] Over the past two decades, LLNL generated an extensive data base on laser damage and conditioning in KDP and DKDP crystals.[2] While the damage thresholds have improved over time — primarily in response to better filtration of growth solutions[3] — they are still far below what is expected from the band structure of the perfect crystal. Thus these empirical studies have shown that damage in KDP, like the other NIF optical materials, is caused by extrinsic defects. The purpose of this project was to perform the basic science needed to understand the process of damage or conditioning and identify the defects responsible for damage. In addition, we sought to develop time resolved spectroscopy and imaging tools that would be generally applicable to investigations of laser-materials interactions.

Results

Most of the results of this project are summarized in the attached publications. *"Temperature and spectral investigations of bulk KDP below damage using 355nm laser irradiation"* presents results showing that light emission occurs during laser damage of KDP. This emission is indicative of internal temperatures in excess of 1000K. *"Raman scattering investigation of KDP subsequent to high fluence laser irradiation"* shows that, although high fluence laser light generates large numbers of electronic defects, such high temperatures must be confined to regions with diameters of less than 100 nm. The results required the presence of either sub-micron, inclusions of highly-absorbing species or, at a minimum, sub-micron clusters of such species. In both cases, we hypothesized that, during laser irradiation, localized heating due to absorption by the inclusion results in its vaporization, followed by damage to the crystal through local melting as well as fracture from thermal stresses. Low fluence laser conditioning should then reduce the probability of damage by heating the inclusions to temperatures below the vaporization point. This increases their solubility and diffusivity so that they become more dispersed and less absorbing. Likely candidates for the damage precursors include transition metal phosphates which are particularly suspect because they are highly insoluble, present in the starting materials, and have high UV absorption coefficients.

"Observation of photoexcited emission clusters in the bulk of KDP and laser conditioning under 355nm irradiation" and *"Microscopic fluorescence imaging of bulk defect clusters in KDP crystals"* describes the results of our attempts to use micron scale images of fluorescence to identify these precursors to damage. The results showed that KDP crystals contained large numbers of fluorescently active defects and defect clusters and that the density of these defects was correlated with the impurity levels in the crystal. *"Investigation of optically active defect clusters in KDP under laser photoexcitation"* and a manuscript in preparation show that the intensity of clusters as well as their number is diminshed by orders of magnitude upon irradiation at 355nm. In other words, these defects have all of the characteristics of damage precursors: they are submicron in size, they consist of species that absorb at 355nm, and they exhibit conditioning. However, no correlation between the location of these defects and subsequent damage was observed.

The spectrum of light emitted during damage should contain the signature of the damage precursor. Unfortunately, the spectrum of the emitted light collected on the micron timescale in our early studies did not represent that of the original damage "fireball" because it was reabsorbed and remitted by defect states associated with impurities in the crystal. In response we developed emission spectroscopy with nanosecond time resolution to look for atomic emission lines during damage. While the spectra revealed high temperature radiation from the bulk, no clear atomic emission signatures were seen above the bright background.

Our final technique for identifying the composition of damage precursors used direct chemical analysis of damage sites. Through careful chemical etching, we brought sites that had been laser damaged near the surface of a crystal. These sites were then investigated by ion milling through them and performing secondary ion mass spectrometry on the ejecta. These measurements revealed sub-micron regions that were rich in Ca, Fe, Cr, and Cu. Some of these species, particularly Fe, are strong UV absorbers when in the form of phosphates. These measurements provide the strongest evidence to date that metal phosphate particles or metal ion clusters serve as the precursors to damage.

Because of the importance of particles and impurities in laser damage of KDP, , we investigated the growth of KDP in the presence of Fe, Cr and Al using *in situ* atomic force microscopy. The results are described in the attached publications *"Recovery of surfaces from impurity poisoning during crystal growth"* and *"A comparison of growth hillock structure and step dynamics on KDP {100} and {101} surfaces using atomic force microscopy"*. They provide the first microscopic picture of impurity-step interactions. They show that growth of KDP surfaces in impurity bearing solutions occurs on close bunches of elementary steps, known as macro-steps, while individual elementary steps are immobilized by the impurities. Based on a simple physical model we were able to derive a characteristic time for impurity adsorption. While we did not observe direct incorporation of foreign particles, because macrosteps are known to lead to anomalous and inhomogeneous incorporation of impurities, these results suggest that macrosteps play an important role in determining the distribution of impurities and their clusters that are the likely source of laser damage.

References

1. J.D. Lindl, Phys. Plasmas 2 (1995) 3933 ; J.A. Paisner , *National Ignition Facility, Conceptual Design Report —Executive Summmary*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-PROP-117093 ES (1994).

2. Rainer, F. Atherton, L.J., De Yoreo, J.J., Laser damage to production- and research grade KDP crystals, in: H.E. Bennett et al., eds.,Laser-induced damage in optical materials:1992,*SPIE* **1848**, 46 (1992).

3. K.E. Montgomery and F.P. Milanovich High-laser-damage-threshold potassium dihydrogen phosphate crystals, *J. Appl. Phys.* **68**, 15 (1990).

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